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Method for measurement of radon diffusion and solubility in solid materials

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ABSTRACT

Summary: In order to study the permeation i.e. the diffusion and solubility of radon gas in biological material, a new setup was constructed and a novel analysis was applied to obtain diffusion and solubility coefficients. Thin slabs of solid materials were installed between detector housing and the surrounding radon exposure chamber of 50 Ls volume. In this setup radon can diffuse through thin test samples into a cylindrical volume of 5 mm height and 20 mm diameter and reach an α-particle detector. There the 5.49 MeV α-decay of the penetrating radon atoms is measured by a silicon surface barrier detector. The time dependent activities inside the small detector volume are recorded after injection of a known radon activity concentration into the outer chamber.

Analyzing the time behavior of the integral α-activity from radon in the small vessel, both, the diffusion coefficient and solubility of the test material can be determined, based on a new mathematical model of the diffusion process concerning the special boundary conditions given by the experimental setup. These first measurements were intended as proof of concept for the detection system and the data analysis. Thin polyethylene foils (LDPE) were selected as material for the diffusion measurements and the results were in agreement with data from literature. In further measurements, we will concentrate on biological material like bone, fat and other tissues.

1. Introduction

Radon as a naturally occurring radioactive noble gas has an important impact on human life: approximately one half of the annual radiation dose caused by natural sources is due to radon [1]. Additionally almost 10% of all lung cancer deaths in the EU are attributed to radon exposure [2]. On the other hand, radon is also used in medicine, for therapy of inflammatory diseases [3]. In a large consortium, the genetic risk and the anti-inflammatory effect of radon exposure to humans are studied in molecular, cellular and animal experiments and patient studies are performed. Basic goal of the biological research in the GREWIS project (Genetic risk and anti-inflammatory effects of ionizing radiation) is to acquire more profound knowledge of the diffusion and solubility of ^{222}Rn and its decay products.

In a biological organism the activity concentrations of radon and consequently its decay products seem not to be homogeneously distributed in the various compartments yielding an inhomogeneous dose to different organs [4]. According to experimental data, especially the radon solubility differs between fatty tissues and tissue containing more water, like muscle [5]. Because of the lack of systematic data for biological samples, we developed an experimental set up for measurements of diffusion, solubility and permeation of radon in various materials.

In previous experiments [6], we used γ-spectroscopy to determine the radon content in thick specimens after radon exposure in an especially designed chamber. There, the radon activity concentration in the sample is determined by the γ-decay of the radioactive decay products ^{214}Bi and ^{214}Pb. The course of the measured activities of these isotopes depends primarily on the half-lives in the order of 20–30 min and secondly on the radon diffusion out of the samples during measurement. In the given setup, it was not possible to determine the diffusion coefficient independently. This is why it was treated as a variable fit parameter in our evaluation model, yielding a high uncertainty.

Consequently, we decided to establish an additional method, measuring both, the diffusion and the solubility in different materials directly. To understand the effect of radon exposure to mankind, the knowledge of transport dynamics of the diffusion process through the tissue, but also the absolute radon concentrations in various critical organs are important. The time constants for diffusion are in the range of minutes, thus in competition to the biological washout through the

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vascular and the lymphatic system in the body. On the other hand, the solubility of radon in the used organic samples is of the same interest for its effectiveness in the body, because solubility scales the concentration in the organs. Consequently, we designed an experiment where both parameters – the diffusion coefficient and the solubility - can be determined simultaneously and in the same sample.

Previous papers in the field of radiation research [7–10] focus mainly on the determination of the diffusion coefficients in radon proof materials or radon diffusion through porous substance. The commonly used experimental setup consists of two chambers between which the test-membrane is fixed. Then radon is flushed into one of the chambers and the change of the radon activity concentrations in both chambers is measured. Another method to measure the diffusion is to wait until the radon activity concentration in both chambers has reached steady-state conditions. Typical measurement times vary from days to weeks [11,12].

For the analysis of experimental data, the diffusion coefficient is calculated according to Fick's law, from the time-dependent course of the radon activity concentration under non-stationary conditions. In general, data for diffusion coefficients given in literature have to be considered carefully, as in some studies the authors obtain data for the permeability but label them diffusion coefficients, which can cause confusion [13]. The determination of permeability only is not sufficient for the understanding of radiobiological effects in various organs of the human body, as mentioned above.

It is noteworthy, that in the research field of permeation and diffusion for non-radioactive gases, papers describe the experimental determination for both, diffusion constant and permeability. For instance, Nguyen et al. [14] found an approximate method that is discussed below in Section 2.2.4.

The diffusion coefficient D and the permeability P are closely connected by the following relation,

$$P = S \cdot D \tag{1}$$

where the proportionality factor S is the solubility of the permeant gas in the sample. The exact definition for P and D are given in Section 2.2, but if the solubility is not known, P cannot be determined from D and vice versa.

The purpose of our experiments is to measure both D and S for the diffusion and solubility of radon in biological materials, like organic tissue samples or fat slices, where no published data are available. We have developed a technique where radon can diffuse, in a reasonable time, from a large vessel through thin slabs of test samples into the volume of a much smaller vessel until the partial pressure in both vessels are equalized. During diffusion time, a surface barrier detector measures precisely the increase of radon activity concentration in the small vessel by recording the energy spectra of the α-decays. The time dependent integral activity of the ^{222}Rn α-line delivers the information for D and S.

Therefore, a mathematical model is introduced, describing the diffusion process in that specific setup with its special boundary conditions. The equations were numerically solved for a wide range of parameters for S and D. These numerical solutions could be fitted in good agreement with an analytical approximation of the solution (quasi analytical solution).

First measurements serve as proof of concept for this technique. For these experiments, we used standard polymers, like polyethylene foils, enabling more defined conditions compared to organic samples and a more extended comparison to data from literature.

2. Materials and methods

2.1. Experimental setup

The general structure of the experimental setup for the diffusion measurements is shown in Fig. 1. All experiments are performed in a radon exposure chamber, having a volume V_0 of 50 liters. The chamber has been constructed for the exposure of mammalian cells and small animals under therapeutic conditions, as used in radon galleries, and is described in detail in [6]. The ^{222}Rn is produced in a ^{226}Ra source and uniquely injected (at time t = 0) into the volume of the large compartment with adjustable activity concentration, ranging up to 600 kBq/m^3. A constant and homogenous partial pressure p_0 of radon is quickly reached after a few seconds. For the measurements, the test samples between a few μm and 10 mm can be mounted in a small housing, which is then located inside of the exposure chamber (large compartment). Therefore only radon gas that penetrates the test sample is detected by its α-decay. The α-detector is a silicon surface barrier detector with an active area of 450 mm^2, and a minimum depletion depth of 500 μm (ORTEC model U-019-450-500). The resolution at an α-energy of 5.486 MeV is 19 keV FWHM in vacuum. In the gas filled volume above the detector the energy resolution is degraded because of the different path lengths, as seen in Fig. 2. The two Po nuclei with energies at 6 and 7.7 MeV are deposited directly on top of the detector while the decay of α- emitters in gas phase produce more complex spectra (see Fig. 2).

The standard electronics consist of an amplifier (IN 7243 E), multichannel analyzer and an analog digital converter (Aspec 927). Usually, the spectra are recorded over a few hours with an interval length of 30 s. In these spectra, after background subtraction, the region of interest, i.e. the energy range of the Rn α-particles, is analysed.

After injection, the radon can reach the detector only when entering through the inlet and diffusing through the sample (thickness L) into the small downstream compartment (height 5 mm, radius 10 mm) with a volume $V \ll V_0$. The partial pressure of the penetrating radon is measured with an α-detector below the downstream compartment. Because α-particles have a range of only a few μm in solid materials, we assume that all recorded α-particles emitted by ^{222}Rn originate in this downstream compartment and not in the test material. The partial pressure of the radon at the upper surface of the test sample (p_0) is equal to the partial pressure in the whole large compartment. The partial pressure p(t, x) of radon in the sample is considered to be a function of the x-direction (perpendicular to the sample) and time. At the lower surface, the partial pressure p(t, L) has the same value as in the whole downstream compartment.

Because of the energy loss in the gas atmosphere above the detector, the α-line is broadened towards lower energies and generates a radon continuum. For thick samples, the α-decay in the volume V_0 above the test samples is not recorded in the detector when the range of the various α-particles is shorter than the thickness of the sample. In Fig. 2, the continuum of ^{222}Rn under a polymer foil with 50 μm thickness is shown together with the polonium lines and a simulated spectrum. The polonium lines are not widened, because these isotopes are heavy metal atoms and aggregate at the surface of the detector. Accordingly, those lines can be used for energy calibration.

In order to prove the plausibility of the measured spectra, a simple simulation was performed. The simulation is intended to reproduce the shape of the ^{222}Rn α-spectra, which are formed by the geometric boundary conditions and the corresponding tracks in the volume above the α-detector. The simulation uses tables of α-particle ranges in air [15] and assumes a homogenous ^{222}Rn activity concentration in the cylinder volume above the detector and an isotropic α- emission. For the α-particles starting at a certain position in the cylinder and with a certain direction angle, the path length up to the detector surface and the corresponding energy loss in air is calculated by the range tables. The remaining energy produces the signal perceived by the detector. Additionally, the energy loss straggling in air was included and a normal distributed fluctuation of σ = 72 keV from the noise of the detector electronics was assumed. The integration over the cylinder volume and the full solid-angle (for the α-particle directions) finally gives the simulated spectra. In Fig. 2 the simulated energy spectrum is compared with the measured spectrum, which shows a qualitatively